

Comment on: "Ferroelectricity in spiral magnets"

There is much interest in the physics of materials that show a strong coupling between magnetic and electric degrees of freedom. In a recent paper by Mostovoy [1], a theory is presented that is based on symmetry arguments and leads to quite general claims which we feel merit some further analysis. In particular, Mostovoy concludes that *spiral magnets are, in general, ferroelectric*.

We argue that this conclusion is not generally valid, and that the symmetry of the unit cell has to be taken into account by any symmetry-based magneto-electric coupling theory. In an attempt to avoid further confusion in the search of new multiferroic materials, we identify in this Comment some of the necessary symmetry properties of spiral magnets that can lead to ferroelectricity.

We take the example of the ferroelectric phase of TbMnO_3 , where the magnetic structure is incommensurate along the crystallographic b-axis, and contains Mn^{3+} moments along the b-axis and c-axis that belong to two different irreducible representations Γ_3 and Γ_2 , respectively [2]. The symmetry of the magnetic structure is described by the direct product of Γ_3 and Γ_2 . Fig. 1a shows that this magnetic structure breaks inversion symmetry and a mirror plane in the ab plane, thereby allowing a ferroelectric polarization $\mathbf{P} \parallel \mathbf{c}$ (but not along other directions) [2]. We reached similar conclusions for multiferroic $\text{Ni}_3\text{V}_2\text{O}_8$ [3, 4].

Now consider a hypothetical magnetic structure for TbMnO_3 that is a magnetic spiral with Mn^{3+} moments still in the crystallographic bc-plane, but with the c spin component belonging to Γ_4 instead Γ_2 . This structure is also a bc-polarized spiral structure, albeit with different phase relations between some of the nearest-neighbors compared to those in the experimentally observed magnetic structure. The symmetry of this structure is given by the direct product of Γ_3 and Γ_4 . Since the relevant magnetic structure is odd under both m_{xy} and m_{yz} and even under 2_y it can not support a polar axis: the ferroelectric moment must be zero even though inversion symmetry is broken. In contrast, according to Eq. 5 of Mostovoy [1], his theory would predict a ferroelectric moment along the c-axis.

Mostovoy's theory may also wrongly predict the direction of electric polarization in some materials. Consider a hypothetical magnetic structure that is a spiral modulated along the b-axis, and where the moments along the b-axis belong to Γ_3 and the moments along the a-axis belong to Γ_2 . According to Mostovoy's theory, this spiral structure supports $\mathbf{P} \parallel \mathbf{a}$. However, as shown above, a magnetic spiral structure belonging to Γ_3 and Γ_2 can only support $\mathbf{P} \parallel \mathbf{c}$, irrespective of the plane of spin rotation.

In conclusion, we do not agree with Mostovoy's statement (which most of his paper is based on) that *incommensurate spin-density-wave states are largely insensitive to details of crystal structure and can be described by a*

continuum field theory of the Ginzburg-Landau type. The examples that we present in this Comment show that the symmetry properties of the crystal lattice and of the magnetic order play a crucial role in phenomenological description of magneto-electric coupling [2, 3], and that the continuum symmetry approach that Mostovoy proposes leads to misleading predictions. Further we note that an additional virtue of dealing with the symmetry of representations [2, 3, 4] is that one sees immediately that perturbations within the representations that lead to magnetic components in addition to either the nonferroelectric collinear structure or to the ferroelectric spiral do not change the symmetry. To reach the same conclusion within Mostovoy's formulation requires additional analysis.

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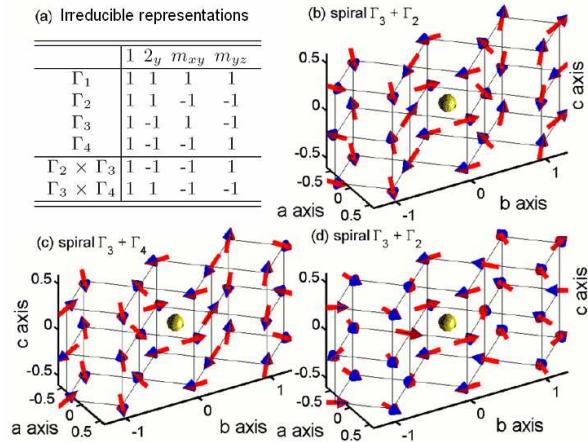


FIG. 1: (a) Magnetic irreducible representations of the Mn^{3+} sites in TbMnO_3 . (b) Inversion-symmetry breaking spiral phase that was found experimentally and leads to ferroelectricity. (c) Hypothetical inversion-symmetry breaking spiral structure that does not allow ferroelectricity (the yellow sphere indicates the broken inversion center). (d) Hypothetical inversion-symmetry breaking spiral structure that allows $\mathbf{P} \parallel \mathbf{c}$, but for which Mostovoy's theory predicts $\mathbf{P} \parallel \mathbf{a}$.